

Attorney Docket No.: OGOSH53USA

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Application No. : 10/595,660 Confirmation No. 4264  
Applicant : Yuichiro Shindo  
371 Filed : May 3, 2006  
Art Unit : 1733  
Examiner : Jessee Randall Roe  
Customer No. : 00270  
Title : HIGH PURITY HAFNIUM, TARGET AND THIN FILM  
COMPRISING SAID HIGH PURITY HAFNIUM, AND  
METHOD FOR PRODUCING HIGH PURITY HAFNIUM

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**APPEAL BRIEF**

Sir:

This is an Appeal Brief submitted in accordance with 37 CFR §41.37. A Notice of Appeal was timely filed on February 14, 2012, and a Petition for a One Month Extension of Time is being submitted herewith to extend the deadline for submitting the Appeal Brief from April 14, 2012 to May 14, 2012. The appeal is taken from a FINAL rejection issued on November 14, 2011 for the above identified application.

### Real Party in Interest

The real party in interest is JX Nippon Mining & Metals Corporation.

The named inventor assigned his rights in the application to Nikko Materials Co., Ltd. via assignment recorded in the U.S. Patent and Trademark Office on May 4, 2006, reel/frame: 017851/0062. A name change document recording a name change from Nikko Materials Co., Ltd. to Nippon Mining & Metals Co., Ltd. was recorded in the U.S. Patent and Trademark Office on November 29, 2006, reel/frame: 018560/0126. More recently, Nippon Mining & Metals Co., Ltd. was merged into Nippon Mining Holdings, Inc. (merger document recorded in the U.S. Patent and Trademark Office on October 8, 2010, reel/frame: 025115/0062) and thereafter, the name of Nippon Mining Holdings, Inc. was changed to JX Nippon Mining & Metals Corporation (name change document recorded in the U.S. Patent and Trademark Office on October 12, 2010, reel/frame: 025123/0358).

### Related Appeals and Interferences

There are no known prior or pending related appeals, interferences or judicial proceedings.

### Status of Claims

Claims 1, 2, 9-11, 14, 15, 18 and 19 are rejected.

Claims 3-8, 12, 13, 16 and 17 are canceled.

Appellant appeals the final rejection of claims 1, 2, 9-11, 14, 15, 18 and 19.

Status of Amendments

No amendment has been filed by the Appellant or entered by the Examiner in the above referenced application since the Final Office Action dated November 14, 2011.

Summary of Claimed Subject Matter

Independent claim 1 is directed to a hafnium material having a purity of 4N (99.99wt%). (See page 5, lines 27-29, of the present application, as filed.) With respect to the above stated purity, the contents of zirconium and gas components within the hafnium material are excluded from consideration as impurities for purposes of determining the purity level of the hafnium material which is conventional practice in the industry for hafnium. (See page 5, lines 27-29, of the present application, as filed.) The hafnium material has a content of oxygen of 40wtppm or less, a content of sulfur of 10wtppm or less, a content of phosphorus of 10wtppm or less, and a content of zirconium of 0.1wt% (1,000wtppm) or less. (See page 5, lines 23-26, of the present application, as filed.)

Independent claim 2 is directed to a sputtering target consisting of high purity hafnium having a purity of 4N or higher excluding zirconium and gas components, an oxygen content of 40wtppm or less, a sulfur content of 10wtppm or less, a phosphorus content of 10wtppm or less, and a zirconium content of 0.1wt% or less. (See page 5, lines 23-32, of the present application, as filed.)

Independent claim 11 is directed to a thin film deposited on a substrate. (See page 5, lines 30-32, of the present application, as filed.) The thin film consists of high purity hafnium having a purity of 4N or higher excluding zirconium and gas components, an oxygen content of

40wtppm or less, a sulfur content of 10wtppm or less, a phosphorus content of 10wtppm or less, and a zirconium content of 0.1wt% or less. (See page 5, lines 23-32, of the present application, as filed.)

None of the claims includes a means-plus or step-plus function permitted by 35 USC §112, sixth paragraph.

#### Grounds of Rejection to be Reviewed on Appeal

Claims 1, 2, 9-11, 14, 15, 18 and 19 stand rejected under 35 USC §103(a) as being obvious over U.S. Patent Application Publication No. 2003/0062261 A1 of Shindo published April 3, 2003.

Claim 1, 18 and 19 stand rejected under 35 USC §103(a) as being obvious over the October 1990 publication of G.T. Murray et al. titled “Preparation and Characterization of Pure Metals” in the ASM Handbook, Volume 2, pages 1093-1097.

#### Argument

##### A. Claim Rejection – 35 USC §103(a) based on the ‘261 Shindo Reference

###### Claims 1, 2, 9-11, 14, 15, 18 and 19

The cited prior art reference, U.S. Patent Application Publication No. 2003/0062261 A1 of Shindo (hereinafter referred to as the ‘261 Shindo Reference), corresponds to U.S. Patent No. 6,861,030 B2 issued to Shindo. Yuichiro Shindo, the inventor of the present application, is the inventor of the invention disclosed in the cited application publication and above referenced

issued patent. JX Nippon Mining and Metals Corporation is the assignee of the present application and the cited reference.

At the outset, Appellant respectfully submits that the subject matter disclosed in the present application represents an advancement of the art over that disclosed in Appellant's earlier filing (the '261 Shindo Reference). Further, Appellant respectfully submits that the '261 Shindo Reference fails to disclose the present invention and would have failed to render the present invention obvious to one of ordinary skill in the art at the time the present invention was made. Still further, Appellant respectfully submits that the "or less" terminology contained in the '261 Shindo Reference and relied upon for purposes of rejecting the claims of the present application is in error and is being interpreted in a mere conclusory fashion without rational underpinning.

More specifically, Appellant respectfully submits that the refining of hafnium according to the '261 Shindo Reference is different than that of the present invention. In addition and most importantly, it should be understood that the awareness of the importance of the presence of zirconium and oxygen in a hafnium material is distinctly different between that disclosed by the '261 Shindo Reference and that required by the claims of the present application.

Hafnium has an extremely strong affinity for oxygen, and since Zr and Hf are homologous elements, it is extremely difficult to mutually separate Zr content from a Hf material. Thus, the purification of hafnium with respect to zirconium and oxygen content is complicated and extremely difficult to achieve and would not have been perceived as being a trivial or obvious endeavor to one of ordinary skill in the art at the time the present invention was made.

The claims of the present application require zirconium content within the hafnium material to be reduced to an extreme level of 0.1 wt% (1,000wtppm) or less and oxygen content to be reduced to an extreme level of 40wtppm or less or 10wtppm or less.

With respect to zirconium content in a hafnium material, the '261 Shindo Reference provides the following express teachings to one of ordinary skill in the art:

“ ... a large quantity of zirconium is contained in hafnium, and notwithstanding the fact that the separation and refinement between the two is difficult, this may be disregarded since the purpose of use of the respective materials will not hinder overall purpose hereof” [Emphasis Added] (see Paragraph No. 0061 of the '261 Shindo published application); and

“It is extremely difficult to reduce Zr in high purity hafnium ... the fact that Zr is mixed in high-purity hafnium will not aggravate the properties of semiconductors, and will not be a problem.” [Emphasis Added] (See Paragraph No. 0065 of the '261 Shindo published application)”

Thus, in accordance to the above referenced teachings of the '261 Shindo Reference, there is no common sense reason provided by the '261 Shindo Reference to reduce Zr content in a hafnium material to 0.1wt% (1,000wtppm) or less (much less to zero or anywhere close to zero or to 0.1wt%). While the '261 Shindo Reference may teach some reduction of Zr content from a raw material hafnium material, the above provides an express teaching that reduction of Zr content from hafnium is not necessary because there is no problem with such content. Reversal of the rejection is requested for at least reason with respect to the contrary teachings concerning Zr content within a Hf material. In addition, the above sentiment of the '261 Shindo Reference with respect to disregarding Zr content in a hafnium material since it does not create a problem, should be kept in mind throughout all additional arguments provided below.

Thus, while the ‘261 Shindo Reference provides a teaching that Zr content within a Hf material will not aggravate the properties of semiconductors, will not be a problem, will not hinder overall purpose, and can be disregarded, Appellant respectfully submits that the ‘261 Shindo Reference also fails to disclose a refining step of subjecting an ingot to deoxidation with molten salt which is the process step that makes possible the reduction of oxygen in the hafnium material to 40wtppm or less, or 10wtppm or less, as required by the claims of the present application. (For example, see page 5, lines 21-26, of the present application, as filed.) With respect to oxygen content, the ‘261 Shindo Reference discloses the following:

“Although it is difficult to reduce the content of gas components such as oxygen and carbon, it is still possible to obtain high-purity zirconium having a content of gas components considerably lower in comparison to the raw material, that is to be less than 1000ppm.” (See Paragraph No. 0063 of the ‘261 Shindo published application.)

Of course, the same holds true for hafnium which is an element that has a strong bond with oxygen and thereby makes the reduction of oxygen from a hafnium material extremely difficult.

In rejecting the claims of the present application based on the ‘261 Shindo Reference, the use and meaning of the phrase “or less” is interpreted by the Examiner to mean a range extending to zero (0.0wt%; 0wtppm). Appellant submits that this is an error. Rejections on obviousness grounds cannot be sustained by mere conclusory statements; instead, there must be some articulated reasoning with some rational underpinning to support the legal conclusion of obviousness. Here, interpreting “or less” with respect to Zr or O content within a Hf material to mean and extend to 0.0wt% or 0wtppm is a mere conclusory statement, is without rational underpinning, and is therefore in error.

In the FINAL Office Action, the claims are rejected on the basis that:

“... the composition disclosed by Shindo (“261) **overlaps** the composition of the instance invention, which is *prima facie* evidence of obviousness.”

Appellant submits this to be an error and respectfully disagrees that the composition disclosed by the ‘261 Shindo Reference overlaps the composition as required by the claims of the present application. Accordingly, Appellant respectfully submits that a *prima facie* case of obviousness cannot be established with the ‘261 Shindo Reference and that this rejection should be reversed for at least this reason.

The ‘261 Shindo Reference discloses a hafnium material having oxygen content of 500ppm “or less” and zirconium content of 0.5wt% (5000ppm) “or less” in a fully-refined hafnium material. Accordingly, one of ordinary skill in the art at the time of the present invention was made is able to acknowledge that the upper limits of O and Zr taught by the ‘261 Shindo Reference for a fully-refined hafnium material are 500ppm and 0.5wt% (5000ppm), respectively. However, one of ordinary skill in the art at the time of the present invention was made was not expressly made aware of the lower limits of O and Zr contents.

One of ordinary skill in the art relying on common sense would clearly understand that a lower limit of content of O and Zr in a hafnium material will always exist and that it will be based on the refining method used. The change in the "free energy" of a metal material upon introducing an arbitrary impurity element into an arbitrary metal material crystal lattice will be a finite value (i.e., it will never be infinite). Thus, regardless of the type of impurity element in a metal material, it is virtually thermodynamically impossible to reduce the concentration of the impurity element to zero (0) or even close to zero. This is particularly true for O and Zr contents

in a hafnium material because O and Zr exist in large quantities in hafnium materials and separation of O and Zr from hafnium materials is extremely difficult.

The purity of a metal obtained from the same disclosed purification process will be of a similar purity level. If the lower limit of the analytical value is determined based on analysis of numerous purified samples, it is possible and reasonable to assume based on common sense that such lower limit will not vary greatly from the analytical values disclosed in the Examples, for instance, the Examples disclosed in the ‘261 Shindo Reference. Here, the Examples disclosed in the prior art reference are relevant for showing the lower limit that one of ordinary skill in the art would reasonable expect based on an common sense evaluation. A purification process has its limits and further reduction of impurities is not expected by one of ordinary skill in the art beyond the limits.

Thus, to determine a reasonable and common sense approach to lower limits, one of ordinary skill in the art relying on common sense would almost certainly look to the examples recited in the ‘261 Shindo Reference and would not merely conclude 0wt% or 0wtppm. The ‘261 Shindo Reference teaches that the amount of zirconium in a fully-refined hafnium material is 3500ppm (0.35%) or 2400ppm (0.24%) and the amount of oxygen (as a gas component) in a fully-refined hafnium material is 120ppm. These analytical values comply with, enable and define the “or less” phrase used in the ‘261 Shindo Reference and provide a rational underpinning with respect to an interpretation of the lower limits without improper reliance on conclusory judgment.

Thus, Appellant respectfully submits that it is an error to conclude that the lower limits of ‘261 Shindo Reference “overlap” with the requirements of 40ppm or less O content and

1000ppm (0.1wt%) Zr content required by the claims of the present application. It is reasonable based on the disclosure of the '261 Shindo Reference that the lower limits are 3500ppm or 2400ppm Zr and 120ppm O as defined by the examples. Thus, Appellant submits that there is no overlap with respect to these ranges. Accordingly, Appellant respectfully submits that a *prima facie* case of obviousness cannot be properly established on this basis. Appellant respectfully requests reversal of the rejection for at least this reason.

Further, Appellant respectfully submits that the '261 Shindo Reference fails to provide a motivation for modifying Zr and O content within its hafnium material to the extent required by the claims of the present application. This is primarily because the '261 Shindo Reference teaches that Zr content can be disregarded as discussed above and fails to teach a method step capable of reducing O content to 40wtppm or 10wtppm or less.

Further, the ranges recited by the claims of the present application provide an unexpected result and are critical for providing the result. The concentrations of Zr and O as impurities are reduced in the present invention for purposes of stabilizing the characteristics of a hafnium composition. This in turn yields a result of making the composition useful in electronic products. See: page 2, line 22, to page 3, line 4; page 4, lines 19-21; page 5, line 33, to page 6, line 2; and page 9, lines 4-9, of the present application, as filed. Note that the '261 Shindo Reference expressly teaches that Zr content within a Hf material will not aggravate or hinder the properties of semiconductors and can be disregarded. Thus, one of ordinary skill in the art would not arrive at the present invention based on the teachings of the '261 Shindo Reference.

The "residual resistance ratio" of a material is generally used as a reference for quantitatively representing the impurity content and processing strain concentration existing in a

high purity metal. The composition required of the claims of the present application is able to produce a thin film having a high residual resistance ratio. For example, see: page 3, lines 1-4; page 4, lines 19-21; page 5, line 33, to page 6, line 2; and page 9, lines 4-13, of the present application, as filed. This is an unexpected result relative to the teachings of the '261 Shindo Reference and is critical for the composition to meet the demands for use in depositing the composition as thin film electronic components. Note in Table 4, page 9, of the present application, as filed, the residual resistance ratio increases dramatically after deoxidation treatment with molten salt. With respect to this point, the '261 Shindo Reference fails to disclose or even suggest the residual resistance ratio of its composition, teaches that Zr content can be disregarded, and fails to use a process of deoxidation with molten salt. Thus, the high purity hafnium of the present invention is not simply an achievement of a higher purity in comparison to the hafnium composition of the '261 Shindo Reference, it is also a composition that yields the unexpected results and effects as follows: a composition having a high resistance ratio which is possible to sufficiently meet the demands as a thin film electronic component material.

For all the above reasons, Appellant respectfully submits that the claims of the present application are patentable and are non-obvious relative to the teachings of the '261 Shindo Reference and that the rejection should be reversed.

B. Claim Rejection – 35 USC §103(a) based on the ASM Handbook

Claims 1, 18 and 19

Appellant respectfully submits that the publication in the ASM Handbook has been misinterpreted and fails to make claim 1 of the present application obvious.

The referenced ASM Handbook relates to chemical vapor deposition and discloses that refining is possible with an iodide pyrolysis method. The ASM Handbook, page 1094, provides the following disclosure:

“One of the more popular of the chemical vapor deposition processes is the iodide process, which has been used extensively to purify titanium, zirconium, and chromium (Ref 5). For each of these metals, the starting charge of metal is reacted to form a volatile metal iodide compound, which in turn is thermally decomposed to liberate iodine vapor. The pure metal is allowed to condense onto a suitable heated substrate (glass tubes and wires of the base metal have been used), while the iodine returns to the metal charge to form more iodide compound. Hence, the iodine acts as a carrier of the metal from the charge to the substrate.

In this process, some impurities are always carried over to the vapor phase along with the metal being purified. ... In all cases, the starting metal has a purity of about 99.9%. Chromium has been purified to its highest state to date by this method. Only iron is carried over with these metals to a significant extent. Thus, if a low-iron starting metal is used, the condensed vapor will approach a purity level of 99.999%.

Other metals that have been purified by chemical vapor deposition include hafnium, thorium, vanadium, niobium, tantalum, molybdenum, and many less commercially important metals (Ref 5).” [Emphasis added]

From the above referenced disclosure provided by the ASM Handbook, it is concluded in the FINAL Office Action that the ASM Handbook discloses “purifying metals such as hafnium to a purity approaching 99.999% by chemical vapor deposition when a low-iron starting material would be used.” Appellant disagrees and submits that this is an error. The ASM Handbook does not disclose a level of purity for hafnium and does not disclose Zr content and O content within a hafnium material subject to this treatment.

At the beginning of the above recited first paragraph, Ti, Zr and Cr are metals of which the iodide process is most broadly applied. In the description “...which has been used extensively to purify titanium, zirconium, and chromium (Ref 5) and for “each of these metals, the starting charge ...”, it is clear that the intention of the authors is that the phrase

“these metals” specifically refers to titanium, zirconium, and chromium which are the only metals having been expressly discussed by the authors of the article to this point in the article. Hafnium has not yet been referenced in the article.

The second paragraph recited above discloses typical purity levels for Ti, Zr and Cr (i.e. “these metals”) that are purified with the iodide process. This paragraph of the ASM Handbook discloses that: “In all cases, the starting metal has a purity of about 99.9%.” These “cases” specifically refer to the cases of Ti, Zr and Cr.

The last two sentences of the second paragraph recited above provide the following description:

“Only iron is carried over with these metals to a significant extent. Thus, if a low-iron starting metal is used, the condensed vapor will approach a purity level of 99.999%.” [Emphasis added]

Again, the authors consistently refer to “these metals”, and “these metals” clearly refer back to Ti, Zr and Cr which are the only metals disclosed in the article to this point in the article. A hafnium material has not yet been disclosed in the article to this point. Thus, reference to “these metals” certainly cannot include a hafnium material since a hafnium material has not yet been referenced by the article.

After the description provided in the second paragraph with respect to “these metals” and a purity level approaching 5N, the ASM Handbook provides the following description:

“Other metals that have been purified by chemical vapor deposition include hafnium, thorium, vanadium, niobium, tantalum, molybdenum, and many less commercially important metals (Ref 5).” [Emphasis added]

A reference to purifying hafnium is included in the recitation of “other metals” and not in connection with the recitation of “these metals”. Accordingly, one of ordinary skill in the art

learns from the ASM Handbook that a CVD method can be applied to “other metals” such as hafnium; however, the ASM Handbook completely fails to teach, in any way, the purity level of Hf that can be achieved with the CVD method or the applicability of the iodide process to Hf. Zr and O contents are also not disclosed for a hafnium material.

Further, the ASM Handbook specifically references a “Ref. 5” in the above paragraphs. From an inspection of “Ref. 5” (of record in the present application), the purity of Hf subject to the iodide process is 98.92 to 99.22% and the oxygen content is 140 to 500ppm. These values are significantly different from that required by independent claim 1 of the present application and clearly do not “overlap” the requirements of the claims of the present application. Accordingly, Appellant respectfully submits that a *prima facie* case of obviousness has not been established with the ASM Handbook.

Accordingly, for all the above reasons, Appellant respectfully submits that the ASM Handbook has been misinterpreted, that it is an error to conclude that the ASM Handbook discloses a hafnium material “overlapping” the requirements of claim 1 of the present application, and that a *prima facie* case of obviousness cannot be provided by the ASM Handbook. Accordingly, Appellant respectfully requests a reversal of the rejection.

### Summary

For the reasons stated above, it is submitted that the final rejection of claims 1, 2, 9-11, 14, 15, 18 and 19 should be reversed.

Payment of \$620 for the required fee under 37 CFR §41.20(b)(2) is charged to our deposit account No. 08-3040. Please charge any deficiency in the fee submitted for this brief to our deposit account 08-3040.

Respectfully submitted,  
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Enclosures:

(a) appendix with copy of claims on appeal

## CLAIMS APPENDIX

### COPY OF CLAIMS INVOLVED IN THE APPEAL

Claim 1 (original): High purity hafnium having a purity of 4N or higher excluding zirconium and gas components, an oxygen content of 40wtppm or less, a sulfur content of 10wtppm or less, a phosphorus content of 10wtppm or less, and a zirconium content of 0.1wt% or less.

Claim 2 (previously presented): A sputtering target consisting of high purity hafnium having a purity of 4N or higher excluding zirconium and gas components, an oxygen content of 40wtppm or less, a sulfur content of 10wtppm or less, a phosphorus content of 10wtppm or less, and a zirconium content of 0.1wt% or less.

Claims 3-8 (canceled).

Claim 9 (previously presented): A sputtering target according to claim 2, wherein said oxygen content is 10wtppm or less.

Claim 10 (previously presented): A sputtering target according to claim 9, wherein said sputtering target has a body produced by subjecting a hafnium raw material to electron beam melting to form a hafnium ingot, subjecting the ingot to deoxidation with molten salt, and forming a sputtering target from the ingot after said deoxidation.

Claim 11 (previously presented): A thin film deposited on a substrate, said thin film consisting of high purity hafnium having a purity of 4N or higher excluding zirconium and gas components, an oxygen content of 40wtppm or less, a sulfur content of 10wtppm or less, a phosphorus content of 10wtppm or less, and a zirconium content of 0.1wt% or less.

Claims 12-13 (canceled).

Claim 14 (previously presented): A thin film according to claim 11, wherein said oxygen content is 10wtppm or less.

Claim 15 (previously presented): A thin film according to claim 14, wherein said thin film is a sputtered thin film produced by subjecting a hafnium raw material to electron beam melting to form a hafnium ingot, subjecting the ingot to deoxidation with molten salt, forming a sputtering target from the ingot after said deoxidation, and depositing said thin film on the substrate by performing sputtering with the sputtering target.

Claims 16-17 (canceled).

Claim 18 (previously presented): High purity hafnium according to claim 1, wherein said oxygen content is 10wtppm or less.

Claim 19 (previously presented): High purity hafnium according to claim 18, wherein said high purity hafnium is produced by subjecting a hafnium raw material to electron beam melting to form a hafnium ingot and subjecting the ingot to deoxidation with molten salt.

EVIDENCE APPENDIX - none

RELATED PROCEEDING APPENDIX - none